



## Short communication

# Hydrometallurgical process for the recovery of high value metals from spent lithium nickel cobalt aluminum oxide based lithium-ion batteries



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## HIGHLIGHTS

- A leaching process for the recovery of Li, Ni, Co, Al from NCA cathode is developed.
- Hydrochloric acid leads to a higher leaching efficiency than sulfuric or nitric acid.
- Ni and Co separation is carried out by precipitation of  $\text{Co}_2\text{O}_3$ ,  $3\text{H}_2\text{O}$  using  $\text{NaClO}$ .
- Nickel is recovered by nickel hydroxide precipitation.
- A high rate of nickel and cobalt recovery is reached.

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## ABSTRACT

A hydrometallurgical process is developed to recover valuable metals of the lithium nickel cobalt aluminum oxide (NCA) cathodes from spent lithium-ion batteries (LIBs). Effect of parameters such as type of acid ( $\text{H}_2\text{SO}_4$ ,  $\text{HNO}_3$  and  $\text{HCl}$ ), acid concentration ( $1\text{--}4\text{ mol L}^{-1}$ ), leaching time (3–18 h) and leaching temperature (25–90 °C) with a solid to liquid ratio fixed at 5% (w/v) are investigated to determine the most efficient conditions of dissolution. The preliminary results indicate that  $\text{HCl}$  provides higher leaching efficiency. In optimum conditions, a complete dissolution is performed for Li, Ni, Co and Al. In the nickel and cobalt recovery process, at first the  $\text{Co}(\text{II})$  in the leaching liquor is selectively oxidized in  $\text{Co}(\text{III})$  with  $\text{NaClO}$  reagent to recover  $\text{Co}_2\text{O}_3$ ,  $3\text{H}_2\text{O}$  by a selective precipitation at  $\text{pH} = 3$ . Then, the nickel hydroxide is precipitated by a base addition at  $\text{pH} = 11$ . The recovery efficiency of cobalt and nickel are respectively 100% and 99.99%.

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## 1. Introduction

Nowadays, the depletion of resources in fossil fuels and the necessity to reduce environmental impact urges people to develop sustainable energy including renewable energy sources such as: advanced batteries, fuel cells, solar cells or wind energy. Batteries and specifically the LIBs are undoubtedly an alternative to reduce the current dependence to fossil fuel resources. The worldwide consumption of batteries is significant and LIBs are already present on the market of nomadic technologies, such as mobile phones and laptops, but also in hybrid and electric vehicles. However, green

energies involve to consider the life cycle of a product from “cradle to grave”, requiring to pay attention to recycling routes for a safe disposal. Efficient recycling of battery materials is essential for a sustainable battery industry. Moreover, the requirements of the European directive on batteries recycling (2006/66/CE Directive) drives industrials to increase the recycling efficiency of LIBs. Finally, economic, strategic, environmental and safety aspects require to consider batteries recycling.

Li-ion batteries represent an active research field which includes a plurality of systems and components. Currently, most of research works are focused on the development of alternative cathode materials to  $\text{LiCoO}_2$ , mainly due to cost and safety issues and containing other transition metals such as nickel or manganese, to substitute partially or totally the cobalt:  $\text{LiFePO}_4$  (LFP),  $\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$  (NMC),  $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$  (NCA) and  $\text{LiMnO}_2$  (LMO) [1].

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In one hand, pyrometallurgical processes are commonly used in industry for recovering valuable metals in spent batteries with a high efficiency and high productivity [2,3]. However, this type of processes involves a loss of materials, emission of hazardous gases, dust and is high energy consumer. Moreover, the residual alloy requires a hydrometallurgical process to refine it into pure forms (salts, hydroxides, metals). In the other hand, basic hydrometallurgical processes represent an alternative and an opportunity to turn spent batteries into pure metals or metal salts with a low energy requirement but produce salts as by-products.

Despite the large number of studies on hydrometallurgical processes for battery electrodes, very scarce information is available on the NCA material, while it is considered as a promising material [4]. Indeed, its high specific energy and power density, as well as its long life span get the attention of the automotive industry [5]. On a recycling point of view, the possibility to use it in industry scale involves to consider the recycling from the early stages of research.

Therefore, this work is focused on a hydrometallurgical route in order to recover and separate valuable metals (Ni, Co) from NCA powder. Indeed, cobalt oxide is six time more expensive than nickel oxide [6]. The mechanical treatments required in the first step of LIBs recycling such as sorting, dismantling and grinding are not considered in this study.

The first step considered is the acid leaching of a chemically stable material. Usually, hydrometallurgical processes use strong acids to leach materials such as  $\text{H}_2\text{SO}_4$  [7–13],  $\text{HCl}$  [14–17] or  $\text{HNO}_3$  [18]. These acids are investigated for different concentrations (between 2 mol  $\text{L}^{-1}$  and 9 mol  $\text{L}^{-1}$ ) and different temperatures (between 40 and 100 °C). Additionally, operational variables like the solid to liquid ratio (around 2–20% (w/v)) and the leaching time are studied in order to size the recovery process.

The second step deals with the selective recovery of nickel and cobalt from the acid solution. According to literature, several main techniques have already been investigated to recover metals from acid solution, such as:

- the solvent extraction [8,10,15,19–23]
- the resin ion exchange [22]
- the electrochemical techniques [24]
- the selective precipitation [16,18,22,25].

Among them, the precipitation technique is selected, which is the easiest to set up at industry scale. However, the main challenge is to separate efficiently elements which have close electronical structures and therefore similar properties but different values.

In the literature, the separative precipitation of transition metal ions has also been intensively studied by usual methods. Generally, the balance can be shifted toward a precipitation reaction by pH variations or/and the presence of reactive compounds. Thus, it is shown that aluminum, cobalt and nickel can be recovered from spent batteries by neutralization [10,16]. It is also well known that nickel can be precipitated by dimethylglyoxime [16,18] and the cobalt by ammonium oxalate [11]. Nickel and cobalt can also be separated using sodium sulfide [20], cobalt sulfide [11], ozone, Caro's acid or chlorine [22]. But these reagents are expensive and often hazardous. This work is focused on the selective recovery of nickel and cobalt from a cathode material by a selective precipitation. According to the literature, such a recycling process has not been addressed yet from a NCA material. Moreover, the previous works dealing with the separation between nickel and cobalt are based either on a liquid/liquid extraction process [10,15,19,22] for the metal purification or on the use of an organic extracting molecule for a selective precipitation of nickel from solutions presenting a very different Ni/Co ratio that studied in the present work [16,18].

## 2. Experimental procedure

### 2.1. Material and reagents

In the present study, NCA material,  $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$ , supplied by Toda Kogyo is investigated. At first, a chemical analysis of the material is performed by an Induced Coupled Plasma Optical Emission Spectrometer (Agilent Technologies 700 Series ICP-OES) with a complete dissolution in acid (70 (vol.%) of  $\text{HNO}_3$  and 30 (vol.%) of  $\text{HCl}$ ) by a microwave digestion system (Multiwave 3000, Perkin Elmer, Anton Paar).

The particles mean size of NCA powder is 7  $\mu\text{m}$  and the specific surface area measured by BET method is 0.41  $\text{m}^2 \text{g}^{-1}$ .

Chemical reactants used for experiments are reagent grade reagents supplied by Sigma Aldrich.

### 2.2. Material leaching and metals separation by precipitation

At first, experiments are conducted to determine the most efficient leaching conditions by studying the acid nature (hydrochloric, sulfuric and nitric acid), the leaching time (3 and 18 h), the concentration (1, 2, 3 and 4 mol  $\text{L}^{-1}$ ) and the temperature (25 and 90 °C), with the solid to liquid (S/L) ratio fixed at 5% (w/v). The cathode material is leached in a 100 mL one neck glass reactor placed into an oil bath to control the reaction temperature. The solution is stirred at 350 rpm and a vapor condenser is used to reduce the water and acid evaporation.

After leaching, the unleached material is filtered with paper filter (VWR Folded qualitative filter paper, 313, particle retention 5–8  $\mu\text{m}$ ), and washed with 50 mL of distilled water. The concentrations of Li, Ni, Co and Al in the leaching liquor are measured to compute the leaching efficiency by chemical analysis performed with ICP-OES (Agilent Technologies 700 Series ICP-OES). The standard deviation of chemical analysis is measured for all considered elements at 0.02 g  $\text{L}^{-1}$ .

A pH-meter (PHM220 Lab pH Meter MeterLab) is used to control pH during metals extraction process. The filtration and washing of precipitates and the chemical analysis of solutions are performed as described in the leaching process. The quantities of precipitated metals are deduced from the analysis of ions concentrations remaining in solution.

### 2.3. Purity analysis of recovered materials

A purity analysis of recovered materials is conducted by Inductively Coupled Plasma – Optical Emission Spectrometer, after dissolving all precipitates in 4 mol  $\text{L}^{-1}$  hydrochloric acid at room temperature and diluting them in an appropriate concentration with distilled water.

## 3. Results and discussion

### 3.1. Leaching process

The process of corrosion has both thermodynamic as well as kinetic implication. In the case of NCA material, the thermodynamic data from the literature allow to predict the dissolution of nickel, cobalt and aluminum in acidic media [26]. However, on a kinetic point of view, the leaching efficiency results from the energy barrier arising in a rate-determining step that controls the mechanism of corrosion. Thus, in order to evaluate the highest efficient conditions for material leaching, experiments have to be conducted by studying the effect of parameters such as acid nature, concentration, temperature and leaching time with a constant solid to liquid ratio (set at 5% (w/v)).

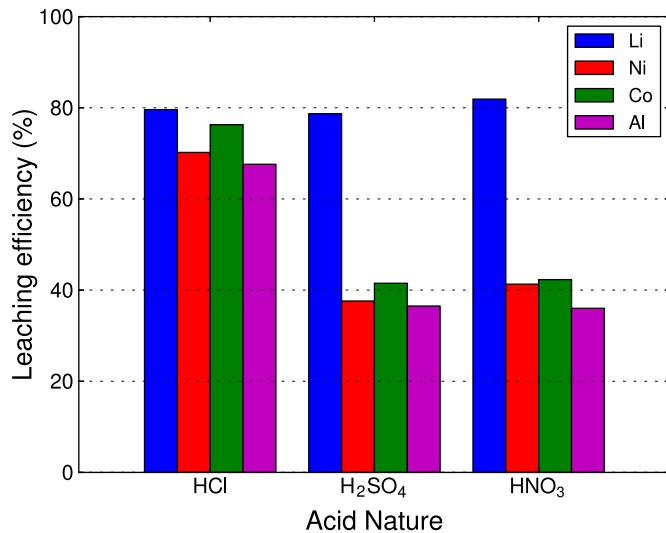


Fig. 1. Acid nature effect on the leaching efficiency of the cathode material ( $C = 2 \text{ mol L}^{-1}$ ,  $t = 18 \text{ h}$ ,  $T = 25^\circ\text{C}$ , S/L = 5% (w/v)).

Fig. 1 shows the effect of acid nature on the leaching for 18 h, in  $2 \text{ mol L}^{-1}$  of acid at room temperature. First of all, these results indicate that the most efficient acid to leach NCA material is the hydrochloric acid, in agreement with previous work [27].

In HCl media, the dissolution rates of Ni, Co and Al are similar. Additionally, the dissolution kinetics of metals from NCA particles show the same behavior in similar leaching conditions, as indicated in Fig. 2. Indeed, except lithium, the dissolution kinetics of other metals are very close in hydrochloric acid. This observation is also valid for sulfuric and nitric acids (Fig. 1). The highest Li concentration observed in the first hours of leaching is probably due to its interfoliar position in the material structure, to move easily between the cathode and the anode during the charge and the discharge of the battery. The 20% of undissolved lithium could be explained by an incomplete dissolution after 18 h and the difficulty for the lithium to diffuse completely due to the inaccessibility to the solution.

On the other hand, a similar kinetic of corrosion of Ni, Co and Al is surprising, while the metals oxidation potential are different, respectively at  $-0.25$ ,  $-0.28$  and  $-1.67 \text{ V}$  vs. SHE. On the base of oxidation potentials, a heterogeneous corrosion can be expected, while homogeneous particles corrosion is observed. Therefore, the

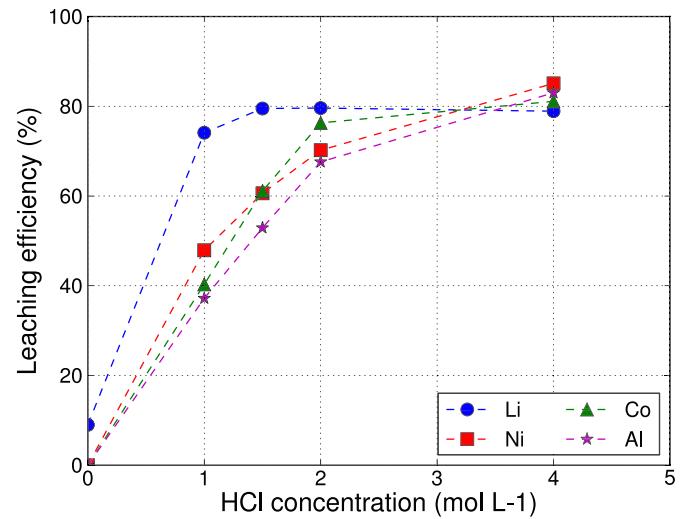


Fig. 3. Effect of the hydrochloric acid concentration on the leaching efficiency of the cathode material ( $t = 18 \text{ h}$ ,  $T = 25^\circ\text{C}$ , S/L = 5% (w/v)).

NCA material has to be considered as a homogeneous material and the dissolution rate of metallic particles is probably controlled by one of them.

Aluminum is known to present a high corrosion resistance due to the formation of a resistive oxide layer which limits the leaching kinetic [28,29]. The oxide layer stability depends on the elements and the pH in solution.

In spite of the high resistance of aluminum in oxidizing environments, it has been largely shown that the presence of aggressive anions like chloride creates localized attacks [30]. This could explain the higher leaching efficiency in hydrochloric acid compared to the others acidic media. Indeed, the electrochemical properties difference between chloride, nitrate and sulfate anions shows a major difference for the aluminum pitting potential. In nitric acid the pitting potential is around  $1.5 \text{ V}$  vs. SCE, while in hydrochloric acid it is around  $-0.75 \text{ V}$  vs. SCE [31]. The potentiodynamic polarization curves in nitric and sulfuric media (exempts of chloride) have both shown an inhibition effect of the corrosion process of aluminum [32,33].

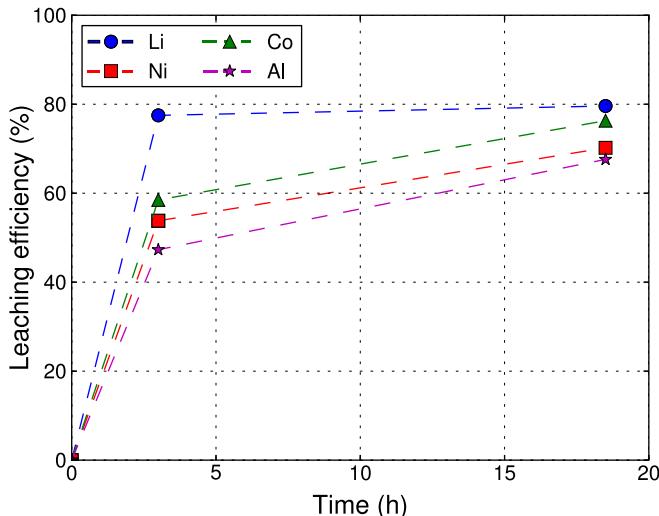


Fig. 2. Effect of the time on the leaching efficiency of the cathode material ( $[\text{HCl}] = 2 \text{ mol L}^{-1}$ ,  $T = 25^\circ\text{C}$ , S/L = 5% (w/v)).

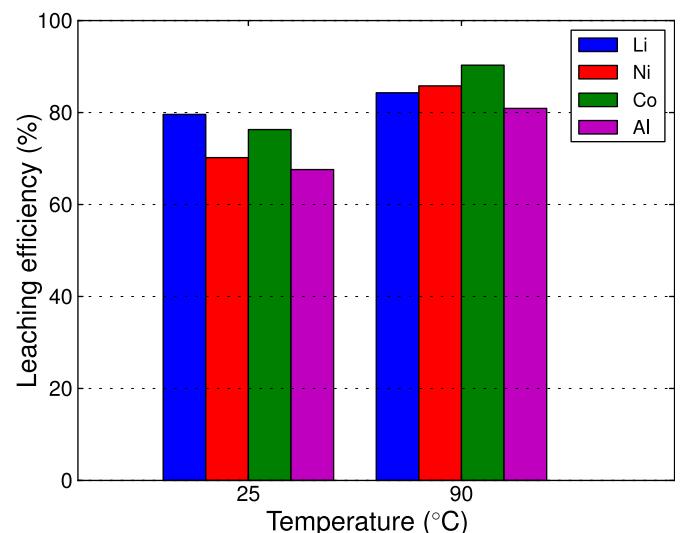


Fig. 4. Effect of the temperature on the leaching efficiency of the cathode material ( $[\text{HCl}] = 2 \text{ mol L}^{-1}$ ,  $t = 18 \text{ h}$ , S/L = 5% (w/v)).

**Table 1**Effect of pH and NaClO/Co ratio on  $\text{Co}_2\text{O}_3$ ,  $3\text{H}_2\text{O}$  recovery.

pH	Ratio NaClO/Co	Co (mol.%)	Ni (mol.%)
2	3	31.5 ± 0.2	3.4 ± 0.1
3		93.8 ± 0.2	4.4 ± 0.1
4		99.8 ± 0.3	7.4 ± 0.1
5		99.5 ± 0.3	18.0 ± 0.1
3	1	17.7 ± 0.3	0 ± 0.1
	2	88.6 ± 0.3	1.5 ± 0.1
	3	93.8 ± 0.2	4.4 ± 0.1

**Table 2**

Effect of pH on nickel recovery.

pH	Ni (mol.%)
6.5	69.7 ± 0.1
11	99.6 ± 0.1
12	99.996 ± 0.051

There is still no general agreement to explain the pit initiation of chloride species [34,35]. However, it is proposed a succession of steps with chloride adsorption, formation of a basic aluminum hydroxychloride salt and dissolution of the oxide. Thus, the concentration in chloride plays a fundamental role in the process of dissolution. Fig. 3 shows the effect of the hydrochloric acid concentration on the leaching efficiency for 18 h, at room temperature. First of all, the overall efficiency is enhanced with the hydrochloric acid concentration. Approximately 80% of lithium, nickel, cobalt and aluminum are leached out when the hydrochloric acid concentration is about 4 mol  $\text{L}^{-1}$ . We note that the same trend was underlined in previous studies for Li, Ni and Co [14,16,17].

Secondly, Fig. 3 shows a logarithmic dependence for the aluminum dissolution in agreement with the observation of Galvele [36]. Indeed, the author observed that the pitting potential shows a logarithmic dependence on chloride ion concentration. The consequence is a shift of the pitting corrosion toward the cathodic potentials due to incorporation of chloride ions into the surface layer [32,33]. Therefore, the surface layer is destabilized with the increase of chloride concentration. Thus, the leaching efficiency is favored with the chloride concentration enhancement.

Others parameters can be studied in order to increase the leaching efficiency. In a kinetic and processing approach, the temperature is a fundamental parameter. For this reason, the temperature effect is studied and is represented in Fig. 4, for a leaching of 18 h in 2 mol  $\text{L}^{-1}$  of hydrochloric acid. As expected, we observe an enhancement of metals dissolution with the temperature. At 90 °C, over 80% of lithium, nickel, cobalt and aluminum are leached out. In the case of Li, Ni and Co, the same trend was already observed in hydrochloric acid solution [16,17].

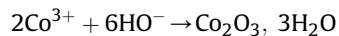
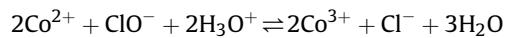
Fig. 4 brings out the similar evolution of the dissolution for Ni, Co and Al. This indicates that under these aggressive acidic conditions, the temperature mainly allows to accelerate the leaching in keeping a homogeneous corrosion mechanism of particles.

From all experiments carried out on NCA material, the most efficient experimental leaching conditions are: 4 mol  $\text{L}^{-1}$  hydrochloric acid, 90 °C, 18 h for 5% (w/v) solid-to-liquid ratio. An

experiment under the above experimental conditions shows that 100% of Li, Ni, Co and Al are leached out, in spite of a possible evaporation of some  $\text{Cl}_2$ .

### 3.2. Selective recovery of cobalt and nickel by oxidative precipitation

From the E-pH diagrams, the separation of Ni and Co is proved difficult due to coprecipitation upon neutralization [26]. When the cobalt and nickel ions are in a same valence state of (+II), the thermodynamic data predict a co-precipitate of  $\text{Co}(\text{OH})_2$  ( $\text{pK}_s = 14.2$ ) and  $\text{Ni}(\text{OH})_2$  ( $\text{pK}_s = 14.7$ ) [37]. However, a selective precipitation is achievable when the cobalt is on a valence state of (+III). In these conditions, the Co(III) precipitation is well dissociated of  $\text{Ni}(\text{OH})_2$  due to a lower solubility product of  $\text{Co}_2\text{O}_3$ ,  $3\text{H}_2\text{O}$  ( $\text{pK}_s = 40.5$ ) [37]. Thus, a selective recovery of valuable metals involves an oxidative precipitation of cobalt. In acidic media, the oxidation of Co(II) occurs at high potential (1.92 V [37]) and requires an oxidant. In this work, the sodium hypochlorite is selected which is a strong oxidant and which has previously proved his efficiency for oxidizing  $\text{Co}^{2+}$  to  $\text{Co}^{3+}$  [25].



A series of experiments is carried out to determine the selectivity of cobalt precipitation. The effect of pH value, and molar ratio NaClO/Co on the  $\text{Co}_2\text{O}_3$ ,  $3\text{H}_2\text{O}$  precipitation is investigated and reported in Table 1.

Table 1 shows that the selective precipitation of cobalt hydroxide is affected by the pH and the ratio of NaClO/Co. It appears that the maximum of cobalt is precipitated at pH = 3. Below this pH, the oxidative precipitation efficiency is strongly reduced. The low content of nickel in  $\text{Co}_2\text{O}_3$ ,  $3\text{H}_2\text{O}$  precipitate is probably due to its entrapment by the solid during solid liquid separation and precipitation during washing with distilled water. Thus, a washing with a solution at pH = 3 is preferable to increase the purity of the cobalt precipitate. Above pH = 3, nickel hydroxide is precipitated significantly.

In parallel, the oxidative precipitation efficiency of  $\text{Co}_2\text{O}_3$ ,  $3\text{H}_2\text{O}$  increases with the NaClO/Co ratio. This effect could be explained by the excess of reactant which shifts the balance reaction to cobalt oxidation.

Regarding nickel recovery, Table 2 indicates a quasi-complete nickel precipitation from pH = 11.

The purity of recovered powders of  $\text{Co}_2\text{O}_3$ ,  $3\text{H}_2\text{O}$  and  $\text{Ni}(\text{OH})_2$  are respectively measured at 90.25 wt.% and 96.36 wt.% (Table 3). The chemical analyses indicate that the major impurities are nickel for  $\text{Co}_2\text{O}_3$ ,  $3\text{H}_2\text{O}$  precipitate and cobalt for  $\text{Ni}(\text{OH})_2$  powder with a low content of aluminum (Table 3). The presence of aluminum in cobalt hydroxide precipitate is probably due to its entrapment by the solid during the filtration and precipitation during the washing step ( $\text{pK}_s = 31.7$  [37]). Its presence in nickel hydroxide precipitate is probably due to precipitation because aluminum hydroxide starts to precipitate at pH = 4 [37]. A washing of the precipitates with a basic solution at pH > 12.4 to solubilize Al in  $\text{Al}(\text{OH})_4^-$  form could

**Table 3**

Purity of cobalt and nickel precipitates (wt.%).

Precipitate	Purity	Li	Ni	Co	Al	Na
$\text{Co}_2\text{O}_3$ , $3\text{H}_2\text{O}$	90.25 ± 0.45	0.49 ± 0.04	5.78 ± 0.23	—	3.31 ± 0.17	0.18 ± 0.17
$\text{Ni}(\text{OH})_2$	96.36 ± 0.04	0.18 ± 0.01	—	2.26 ± 0.03	1.06 ± 0.02	0.13 ± 0.02

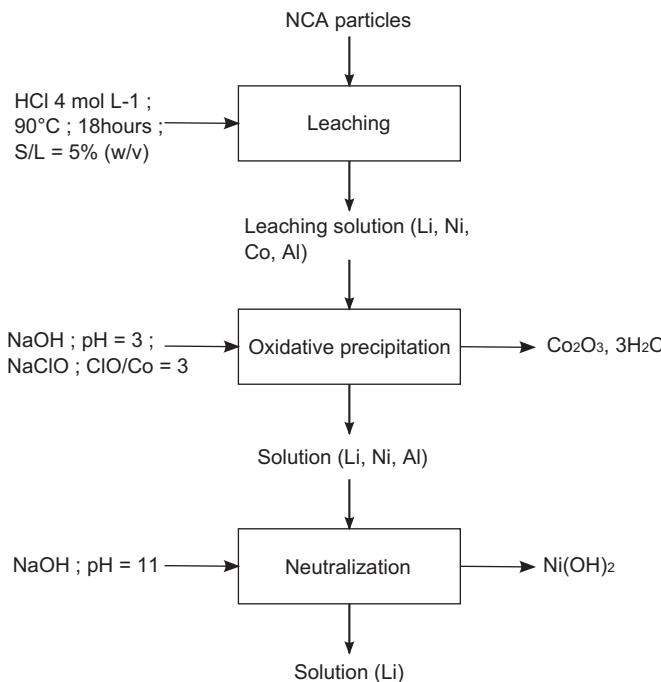


Fig. 5. Treatment process of NCA cathode.

increase the hydroxides purities. Moreover, a more intensive washing could also increase the precipitates purities by decreasing lithium and sodium contents.

On the base of the overall results, a hydrometallurgical flow sheet of treatment process of NCA cathode is defined and presented in Fig. 5.

The recovery of lithium has already been investigated by precipitation as  $\text{Li}_3\text{PO}_4$  or  $\text{Li}_2\text{CO}_3$  [38] with high recovery efficiency (>80%) and good purity.

#### 4. Conclusion

The leaching of NCA cathode has been investigated in sulfuric, nitric and hydrochloric acids. It has been observed that the leaching step is significantly affected by the nature of species with the best leaching efficiency in HCl solution. These differences in acidic media have been attributed to the presence of chloride ions to promote the dissolution. This observation has been ascribed to the ability of chloride ions to destabilize the formation of a surface layer. The optimum leaching conditions have been found to be 4 mol L<sup>-1</sup> hydrochloric acid, 90 °C, 18 h with 5% (w/v) solid to liquid ratio. Under these experimental conditions, all valuable metals of cathode material are leached out.

Based on the recovery material process, it is possible to recover the valuable metals of cathode material from the LIBs. For the cobalt, it has been recovered by oxidative precipitation. The optimum operating conditions of  $\text{Co}_2\text{O}_3$ , 3H<sub>2</sub>O precipitation has been determined for the molar ratio of NaClO/Co of 3 and pH = 3. Regarding the nickel recovery, it is completely precipitated by addition of sodium hydroxide until pH = 11 in a second step.

The experimental results show that the efficiencies of cobalt and nickel recovery are respectively 100% and 99.99%. The purity of cobalt hydroxide and nickel hydroxide are measured respectively at 90.25 wt.% and 96.36 wt.%. These low purities are due to the

presence of nickel in  $\text{Co}_2\text{O}_3$ , 3H<sub>2</sub>O precipitate, cobalt in  $\text{Ni(OH)}_2$  precipitate, and aluminum. The main drawback of recovery and elements separation by hydroxides precipitation is a low purity of recovered materials which require a post treatment to be purified.

Finally, at the end of the process, the leach solution is largely composed of lithium, which could be recovered as a carbonate or phosphate salt [39].

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